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Atmospheric deposition of particulate matter between Algeria and France: Contribution of long and short-term sources

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ABSTRACT

Large worldwide sources of dust, such as the Saharan desert, play a key role in the amounts and composition of atmospheric particulate deposition (APD), but their relative contribution compared to other sources remain unclear. Our study aimed to apportion Saharan, regional, and anthropogenic sources of APD in three sampling along a long transect affected by Saharan outbreaks. We quantified total APD, and analyzed its mineralogical and chemical composition between 2011 and 2012. . Strong markers of Saharan dust, such as large amounts of APD and of a low Al_2O_3:CaO ratio allowed identifying clear periods influenced by Saharan outbreaks. Nitrogen and phosphorus reflected soil affected by agricultural practices, while Cd, Pb, Sb and Sn tracked traffic and industrial sources – as confirmed by enrichment factors. Then, we designed a conceptual model including sensitivity analyses to estimate the contribution of unanalyzed (10–11%, likely chlorites or sulfates), organic (34–41%), anthropogenic (11–22%), mineral-regional (5–25%) and mineral-Saharan (10–38%) matter over our entire study period. Our study shows the rapidly decreasing contribution of Saharan outbreaks – and the decreasing flux of Ca and Mg, from 40 to 0.4 kg.ha^{-1} over the study period between Algeria and France. Yet, Saharan outbreaks were still noticeable in APD at a site 1500 km away. Our study also shows the large relative contribution of organic and anthropogenic sources to APD in the three sampling sites, and their possible influence on nutrient budgets.

1. Introduction

Aeolian or airborne dust largely contributes to the global atmospheric composition with an estimated 2000 Mt emitted into the atmosphere every year (Lawrence and Neff, 2009; Shao et al., 2011). With other compounds, it forms atmospheric particles, which affect health (Brunekreef and Holgate, 2002; Goudie, 2014), climate (Choobari et al., 2014), and ecosystems. In nutrient limited terrestrial ecosystems, such as forests, atmospheric deposition contributes to nutrient inputs, especially when dissolved (Avila et al., 1998; Celle-Jeanton et al., 2009; Lequy et al., 2013b; Meybeck, 1983; Plaisance et al., 1997; Sanusi et al., 1996). Atmospheric particulate deposition (APD) may supply calcium and phosphorus to non-fertilized forests at least as much as weathering fluxes (Lequy et al., 2014a). Monitoring airborne particles and their deposition is thus of primary importance to assess nutrient budgets over terrestrial ecosystems. Major sources of tropospheric airborne dust include the Saharan Desert (Goudie and Middleton, 2006) in which lies the Bodélé depression (Washington et al., 2009) - the largest source on Earth - and other arid regions of Northern Africa, the Arabian Peninsula, Central Asia and China. Lesser sources include Australia, the arid regions of America, and South Africa (Engelbrecht and Derbyshire, 2010).

Saharan dust has often been observed in Europe, following a South-North gradient whether as suspended particles or deposits (Castillo et al., 2017; Marinou et al., 2017; Sicard et al., 2016; Vincent et al., 2016). Spain has witnessed numerous Saharan outbreaks with noticeable changes in the rainfall and dust composition (Avila et al., 1998; Avila and Penuelas, 1999; Escudero et al., 2011, 2006). Saharan dust was collected in the Alps, England, and up to Scandinavia (Bücher and Dessens, 1992; Deangelis and Gaudichet, 1991; Franzen et al., 1994; Telloli et al., 2018). In Northeastern Spain, Saharan outbreaks contributed about 80% of APD (Castillo et al., 2017); in France, regional erosion, rather than Saharan dust, was found to be the main contributor to APD (Lequy et al., 2013a). Nutrient inputs from APD are calculated from deposit flux and its chemical composition, which depends on sources of particles (Kandler et al., 2007; Pey et al., 2009; Plaisance

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et al., 1997; Ridame et al., 1999; Usero and Gracia, 1986). Therefore it is of primary importance to better understand how the distance from large sources such as Saharan desert affects mineralogical and chemical compositions of APD, and the different contributions of regional or local sources. We need to better assess the origin and composition of APD in forests to better assess nutrient inputs and manage forests, especially in a context of global changes and uncertainties about the frequency and intensity of future Saharan outbreaks (Goudie, 2009). APD sampling in forest provides relevant data to better understand forest nutrition.

This study investigates to what extent major long-distance and regional sources, such as Saharan Desert, regional erosion and anthropogenic atmospheric deposition, contribute to APD. To do so, we analyzed the deposition rates, and the mineralogical and chemical compositions of APD in three contrasted sampling sites, from very close to the Sahara desert to more than 2000-km northwards, and deciphered the origin of APD sampled in each site.

2. Material and methods

2.1. Sites description

Three rural sites were monitored in forested ecosystems along a Sahara – Europe axis: in Northern Algeria, in Northeastern Spain and in Northeastern France (Fig. 1). They are located in remote areas with agricultural background.

The Algerian site was located in Tizi Rached (Kabylie) in North Algeria (36°40′N; 04°12′E), at 436 m a.s.l. in the heart of an olive grove, and the vegetation is typical of maquis shrubland. The study site lies in an agricultural region dominated by olive and fig groves, moderate density population cities, and crop fields. The local bedrock is schist and the local soils are classified as Cambisols. The climate is sub-humid with annual rainfall of 826 mm.

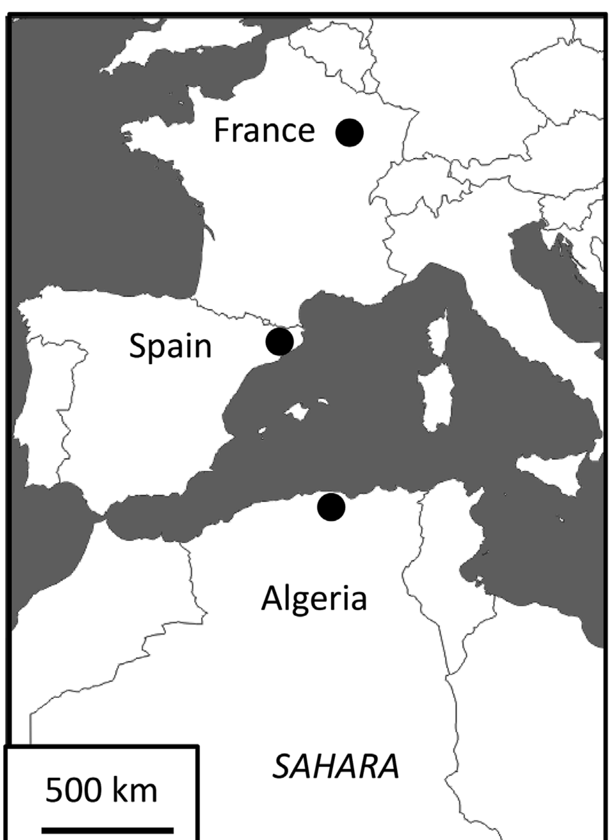

Fig. 1. Localization of the three sampling sites with respect to the Saharan Desert.

The Spanish site was located in La Castanya valley in the natural park of the Montseny mountains (40°46′N; 02°21′E), 40 km north-northeast of Barcelona, and described by Avila et al. (1998). Briefly, the Spanish site is at 700 m a.s.l in the slopes of the valley which is covered by a dense holm oak forest. Southwards lies the pre-coastal depression, where urban, industrial, and agricultural land uses dominate. The site pertains to the Global Atmopsheric Watch and ACTRIS programs, and is equipped with other instruments for the monitoring of the air quality. Concerning its atmospheric quality, the site is considered as a background station (Perez et al., 2008; Pey et al., 2009). The local bedrock is a metamorphic phyllite, with quartz, chlorite, albite, and muscovite as major minerals. Soils are very stony, sandy loam dystric or typic Xerocrepts. The climate is Mediterranean, but the characteristic summer drought is attenuated by summer storms.

The French site was located in Montiers-sur-Saulx in the heart of the beech forest of Montiers in Northeastern France (48°32′N; 05°18′E), and was described in Lequy et al. (2014a). Its elevation is 398 m a.s.l. The landscape includes calcareous plateaus featured by small valleys. This site belongs to the French network SOERE F-ORE-T (long-term observation and experiment system for the environmental research that studies the functioning of forest ecosystems) and to the SOERE OPE (long-term environmental monitoring system) managed by Andra (the French National Radioactive Waste Management Agency). Only small roads and agricultural fields can affect the site as local pollution. The region is rural with a low population density, and the cities nearby the site include only a few hundred inhabitants. Different soils cover the sampling area: principally the calcareous soils (pHw = 5.1), but locally some more acid detrital sediments were encountered (pHw: 4.8). The climate is subcontinental.

2.2. Sampling and sample preparation

2.2.1. Design of the sampling on the field, sample collection, and lab preparation

Passive APD collectors (details hereafter) were placed within olive groves (Algeria) or forests (Spain and France) at 0.2 (Algeria), 0.5 (Spain) and 1.0 (France) km from the vegetation edge. In Montseny, the APD collectors were placed in a clearing at a distance of 300–500 m to the edge of the forest.

Collectors were placed on a 45 m high tower above forest canopy in the French site, and at 2.5 m above ground level in the Algerian and Spanish sites. Four collectors were used in the French site and samples were made after pooling for each sampling period.

Dust collectors and sample preparation were described in Lequy et al. (2014b). Briefly, open field APD samplers consisted in 0.22 m^2 funnels connected to 10 L (Spain) or 20 L (Algeria and France) collecting containers with a nylon sieve to protect samples from coarse organic pollution. A dark protection limited the influence of light in the collection bottles to prevent algal development. In the field, the collection surfaces were cleaned with deionized water and plastic brushes into collection containers. Samples collected in Spain and in Algeria were refrigerated and stored at 4 °C, then sent to our laboratory in France each season. During this delay, some APD dissolution could occur. The French samples were prepared directly after their collection in the field every four weeks. At this step, all the samples were prepared and analyzed in the same laboratory (BEF-INRA Nancy France). Samples containing particles and rainwater are centrifuged during 40 min at 3500 tr. min^{-1}. The particles were transferred in plastic tubes and dried at 35 °C in a ventilated oven and weighted. The protocol recovery was tested from standard minerals and was estimated at 72% (Lequy et al., 2014b). This recovery rate is based on an experiment that mixed coarse, silt and clay fraction of a soil: as such, it represents the average recovery regardless the particle size. Losses were probably larger in finer fractions.